Interaction of a three-level system with two strong fields

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Abstract. General expressions for the steady-state population distribution and complex susceptibilities of a three-level system interacting with two strong near-resonant electromagnetic fields are obtained using a density-matrix formalism. A complete treatment of the steady-state population distribution established in the three-level system, under the action of two strong fields with arbitrary resonance detunings is presented. The criterion for maximal populations of the exciting levels, as well as conditions in which the population of the exciting levels exceeds the population of the ground state are found. The modification of the absorption and emission lines with increasing intensity of the second (probe) field is considered. The emphasis in the present treatment, as distinct from Panock and Temkin, is on the consideration of the arbitrary distribution of the equilibrium populations of the system which exhibits a number of interesting features which have not been previously discussed.

1. Introduction

The interaction of a three-level system with two resonant fields has been extensively discussed in the literature. Starting with the works of Javan (1957) and Kantorovich and Prokhorov (1957), this problem has been investigated by many authors (Hansch and Toschek 1970, Beterov and Chebotaev 1974 and references therein, Shimoda 1976), concentrating their attention on various aspects of the problem, applying different treatments and occasionally a different terminology.

In most cases, however, the response of a three-level system to the action of the resonance fields is considered for the case when one of the fields is a weak probe field or is equal to zero. The treatment of the case, when the second field is also strong, leads to a considerably more complicated problem and only in recent years have some works appeared (Whitley and Stroud 1976, Krainov 1976, Panock and Temkin 1977, Cohen-Tannoudji and Reynaud 1977), in which no restrictions were imposed on the magnitude of the second field.

The paper of Panock and Temkin (1977) comprises a detailed investigation of the emission and absorption lineshapes in a three-level system interacting with two laser fields of arbitrary intensities and resonant detunings, according to figure 1. These authors were the first to find the condition for maximum amplification of the system in the presence of two strong fields. The analysis is carried out by Panock and Temkin for the particular case of equal relaxation times and population distribution $\rho_{22}^0 = \rho_{33}^0 = 0$, where $\rho_{22}^0$ and $\rho_{33}^0$ are the equilibrium populations of levels 2 and 3, respectively.

In the present paper we remove these simplifying assumptions and in § 2 general expressions for susceptibilities and steady-state population distribution are presented.
The steady-state population distribution, established in the three-level system subjected to two simultaneous strong near-resonant fields is presented in § 3. The modifications of the emission and absorption lines with increasing the intensity of the second (probe) field are described in § 4. Here, a special emphasis is given to the system with $\rho_{22} - \rho_{33} \neq 0$.

2. Equations of motion and their solutions

Consider a homogeneously broadened three-level system interacting with two laser fields—an exciting field at the frequency $\omega_0$ close to the transition frequency $\omega_{31}$ between levels 1 and 3 and a Stokes field at the frequency $\omega_s$ close to the transition frequency $\omega_{32}$, as shown in figure 1.

We shall describe the behaviour of the system using a density-matrix formalism. The laser fields are treated classically and the total field is assumed to be:

$$E = \hat{\varepsilon}_0 e^{-i\omega_0 t} + \hat{\varepsilon}_s e^{-i\omega_s t} + cc$$

where the complex amplitudes are independent of time.

The equations of motion for the density-matrix elements can be written in the form (Bloembergen 1965)

$$\frac{\partial \rho_{ij}}{\partial t} + i(\omega_{ij} - i/\tau_{ij})\rho_{ij} = -i/\hbar \sum_k (V_{ik}\rho_{kl} - \rho_{ik} V_{kl})$$

$$\frac{\partial \rho_{ii}}{\partial t} = -i/\hbar \sum_k (V_{ik}\rho_{kl} - \rho_{ik} V_{kl}) + \sum_k (W_{kk}\rho_{kk} - W_{ik}\rho_{ii})$$

where $V_{ik}$ are the matrix elements of the interaction Hamiltonian taken in the dipole approximation: $V = -d \cdot E$. We assume $V_{ii} = 0$, $V_{12} = 0$, i.e. the system does not have a permanent dipole moment and the transition 1–2 is forbidden in the dipole approximation, $W_{ik}$ are the transition probabilities between levels $i$ and $k$, connected with the longitudinal relaxation times $T_{ik}$ by the relation $W_{ik} = \rho_{kk}^{0}/T_{ik}$, where $\rho_{kk}^{0}$ are the equilibrium matrix elements; and $\tau_{ik}$ are the transverse relaxation times.

Under steady-state conditions the solution of (2) can be looked for in the form:

$$\rho_{13} = \sigma_{13} e^{i\omega_{01} t}$$

$$\rho_{23} = \sigma_{23} e^{i\omega_{12} t}$$

$$\rho_{12} = \sigma_{12} e^{i(\omega_0 - \omega_{12}) t}$$

$$\rho_{ii} = \sigma_{ii}.$$
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This substitution reduces the equations of motion to the following algebraic set of equations for the time independent matrix elements $\sigma_{ik}$:

1. \[
(\omega_1 - \omega_0 + i/\tau_1)\sigma_{13} = 1/\hbar (d_{13}\bar{\sigma}^*_0 (\sigma_{33} - \sigma_{11}) + d_{23}\bar{\sigma}^*_3 \sigma_{12})
\]

2. \[
(\omega_2 - \omega_3 - i/\tau_3)\sigma_{32} = 1/\hbar (d_{32}\bar{\sigma}^*_1 (\sigma_{22} - \sigma_{33}) + d_{31}\bar{\sigma}^*_0 \sigma_{12})
\]

3. \[
(\omega_1 - \omega_0 + \omega_3 + i/\tau_2)\sigma_{12} = 1/\hbar (d_{32}\bar{\sigma}^*_1 (\sigma_{13} - d_{31}\bar{\sigma}^*_0 \sigma_{32})
\]

\[
\sum_k (W_{k1}\sigma_{kk} - W_{1k}\sigma_{11}) + i/\hbar (d_{13}\bar{\sigma}^*_0 (\sigma_{13} - d_{31}\bar{\sigma}^*_0 \sigma_{32}) = 0
\]

\[
\sum_k (W_{k2}\sigma_{kk} - W_{2k}\sigma_{22}) + i/\hbar (d_{23}\bar{\sigma}^*_3 (\sigma_{32} - d_{32}\bar{\sigma}^*_3 \sigma_{33}) = 0
\]

After adding the normalisation condition:

\[
\sigma_{11} + \sigma_{22} + \sigma_{33} = 1
\]

one obtains a complete set of equations, which can be solved exactly.

Solving Equations (4a)–(4c) for the non-diagonal elements yields:

\[
\sigma_{13} = \frac{d_{31}\bar{\sigma}^*_0}{\hbar} \left[ \frac{R_{13}}{\Delta_0 + i/\tau_1} + \kappa \left( \frac{R_{13}}{(\Delta_0 + i/\tau_1)^2} - \frac{R_{23}}{(\Delta_0 + i/\tau_1)(\Delta_1 - i/\tau_2)} \right) \right]
\]

\[
\sigma_{32} = \frac{d_{32}\bar{\sigma}^*_3}{\hbar} \left[ \frac{R_{23}}{\Delta_1 - i/\tau_2} + \kappa \left( \frac{R_{13}}{(\Delta_0 + i/\tau_1)(\Delta_1 - i/\tau_2)} - \frac{R_{23}}{(\Delta_1 - i/\tau_2)^2} \right) \right]
\]

where the notations $\Delta = \omega_1 - \omega_0$, $\Delta_1 = \omega_3 - \omega_0$, $R_{ik} = \rho_{ik} - \rho_{kk}$, $\kappa = |d_{31}\bar{\sigma}^*_0/\hbar|^2$ and $\bar{\kappa} = |d_{32}\bar{\sigma}^*_3/\hbar|^2$ are introduced.

These expressions describe the non-diagonal matrix elements in terms of the population differences $R_{ik}$. The latter can be obtained by solving Equations (4d), (4e) and (5) with $\sigma_{ik}$ given by (6). So:

\[
R_{13} = (R_{13}^* - 2(A_0 + V)T_0^* - 2ST^*_0) \left( 1 + 2(A_0 + W)T_0 + 2(A_1 + V)T_1 \right)
\]

\[
+ 2ST_k + A_0(A_1 + V) + S(A_0 + A_1 + V) + W(A_1 + V + S) \right)^{-1} \rho_{11} + T_{12} \rho_{22} + T_{23} + T_{13} \rho_{33} / T_{12}T_{23}
\]

\[
R_{23} = (R_{23}^* - 2(A_0 + W)T_0^* - 2ST^*_0) \left( 1 + 2(A_0 + W)T_0 + 2(A_1 + V)T_1 \right)
\]

\[
+ 2ST_k + A_0(A_1 + V) + S(A_0 + A_1 + V) + W(A_1 + V + S) \right)^{-1} \rho_{11} + T_{12} \rho_{22} + T_{23} + T_{13} \rho_{33} / T_{12}T_{23}
\]

where $A_0$, $A_1$, $W$, $V$ and $S$ can be interpreted as the probabilities for one- and two-photon transitions induced by the fields $E_0$ and $E_k$ (see also Apanasevich 1977)

\[
A_0 = -2 \Im \frac{\bar{\kappa}}{\Delta_0 + i/\tau_1}, \quad A_1 = 2 \Im \frac{\bar{\kappa}}{\Delta_1 - i/\tau_2}
\]

\[
S = -2 \Im \bar{\kappa} \left( \Delta_0 - \Delta_1 + \frac{\bar{\kappa}}{\Delta_1 - i/\tau_2} - \frac{\bar{\kappa}}{\Delta_0 + i/\tau_1} \right)^{-1} \Delta_0 + i/\tau_1 \left( \Delta_1 - i/\tau_2 \right)^{-1}
\]
We have introduced here, the effective relaxation times $\tau$ in the same manner as Clogston (1958).

\[
T_0 = \left[ \frac{1}{T_{21}} \rho_{11}^0 + \frac{1}{2} \left( \frac{1}{T_{12}} + \frac{1}{T_{32}} \right) \rho_{22}^0 + \frac{1}{T_{23}} \rho_{33}^0 \right] \\
\times \left( \frac{1}{T_{12} T_{13}} \rho_{11}^0 + \frac{1}{T_{12} T_{32}} \rho_{22}^0 + \frac{1}{T_{13} T_{32}} \rho_{33}^0 \right)^{-1}
\]

\[
T_1 = \left[ \frac{1}{2} \left( \frac{1}{T_{12}} - \rho_{11}^0 + \frac{1}{T_{13}} \rho_{33}^0 - \rho_{22}^0 \right) \left( \frac{1}{T_{12} T_{13}} \rho_{11}^0 + \frac{1}{T_{12} T_{32}} \rho_{22}^0 + \frac{1}{T_{13} T_{32}} \rho_{33}^0 \right)^{-1}
\]

\[
T_0^* = \frac{1}{2} \left( \frac{1}{T_{12}} \rho_{22}^0 - \rho_{11}^0 \right) + \frac{1}{T_{13}} \left( \rho_{33}^0 - \rho_{11}^0 \right) \\\n\times \left( \frac{1}{T_{12} T_{13}} \rho_{11}^0 + \frac{1}{T_{12} T_{32}} \rho_{22}^0 + \frac{1}{T_{13} T_{32}} \rho_{33}^0 \right)^{-1}
\]

\[
T_s = \left[ \frac{1}{2} \left( \frac{1}{T_{12}} + \rho_{11}^0 + \frac{1}{T_{13}} \rho_{33}^0 + \rho_{22}^0 \right) \left( \frac{1}{T_{12} T_{13}} \rho_{11}^0 + \frac{1}{T_{12} T_{32}} \rho_{22}^0 + \frac{1}{T_{13} T_{32}} \rho_{33}^0 \right)^{-1}
\]

\[
T_i = \frac{1}{2} \left( \frac{1}{T_{12}} \rho_{33}^0 - \rho_{11}^0 + \frac{1}{T_{23}} \rho_{22}^0 - \rho_{11}^0 \right) \\\n\times \left( \frac{1}{T_{12} T_{13}} \rho_{11}^0 + \frac{1}{T_{12} T_{32}} \rho_{22}^0 + \frac{1}{T_{13} T_{32}} \rho_{33}^0 \right)^{-1}
\]

From (3), (6) and the expression of the induced macroscopic polarisation $P = NS\rho(p\rho d)$ (where $N$ is the number of systems per unit volume) we obtained the following form of the susceptibilities at frequencies $\omega_0$ and $\omega_s$:

\[\chi(-\omega_0) = N \frac{|d_{31}|^2}{\hbar} \left[ \kappa \left( \frac{R_{13}}{(\Delta_0 + i/\tau_{13})^2} - \frac{R_{23}}{(\Delta_0 + i/\tau_{32})^2} \right) \left( \Delta_0 - \Delta_1 + i/\tau_{12} + \frac{\tilde{\varepsilon}}{\Delta_1 - i/\tau_{32}} - \frac{\kappa}{\Delta_0 + i/\tau_{31}} \right)^{-1} \right] \quad (10a)\]

\[\chi(\omega_s) = N \frac{|d_{32}|^2}{\hbar} \left[ \kappa \left( \frac{R_{13}}{(\Delta_0 + i/\tau_{31})(\Delta_1 - i/\tau_{32})} - \frac{R_{23}}{(\Delta_1 - i/\tau_{32})^2} \right) \left( \Delta_0 - \Delta_1 + i/\tau_{12} + \frac{\tilde{\varepsilon}}{\Delta_1 - i/\tau_{32}} - \frac{\kappa}{\Delta_0 + i/\tau_{31}} \right)^{-1} \right] \quad (10b)\]

where $R_{ik}$ are given by equation (7).
Expressions (10) and (7) describe the behaviour of the three-level system irradiated by two near-resonant fields in the quite general case, when the intensities and frequencies of the fields are arbitrary as well as the characteristics of the excited system (different relaxation times, dipole momenta and arbitrary equilibrium population distributions). The analysis of the general case meets with serious difficulties because of the rather complex and cumbersome expressions of the $R_{ik}$, but on the basis of the formulae (7) and (10) different physical situations can be considered, choosing the suitable approximations on the relaxation constants. For example, if one assumes that only $W_{31} \neq 0$, $W_{32} \neq 0$ and $\tau_{13} = \frac{1}{2} W_{31}^{-1}$; $\tau_{32} = \frac{1}{2} (W_{31}^{-1} + W_{32}^{-1})$ then $\chi$ and $R_{ik}$ describe the behaviour of the system in which the only damping mechanism is the natural radiative decay (Whitley and Stroud), or if $W_{ik} = (\rho_{ik}^T)/T + \gamma_{ik}$, where $\rho_{ik}^T/T$ and $\gamma_{ik}$ are connected with the thermal and non-thermal collisional processes respectively, one can obtain the results of Osche (1978).

As the emphasis in the present treatment is on the modification of the behaviour of a three-level system caused by the enhancement of the intensity of the Stokes field, further we shall limit our consideration to the particular case of equal relaxation times. Substituting $T_{ik} = \tau_{ik} = \tau$, we can write the imaginary parts of the susceptibilities (10) and the population differences (7) in the form:

$$
\chi''(-\omega_0) = N \frac{|d_{31}|^2}{\hbar} \tau \text{Im} \left[ \frac{R_{13}}{\delta_0 + i} + \kappa \left( \frac{R_{13}}{\delta_0 + i} - \frac{R_{23}}{\delta_0 + i - \delta_0 - i} \right) \left( \delta_0 - \delta_s + i + \frac{\epsilon}{\delta_0 - \delta_s} - \frac{\kappa}{\delta_0 + i} \right)^{-1} \right]
$$

$$
\chi''(\omega_s) = N \frac{|d_{32}|^2}{\hbar} \tau \text{Im} \left[ \frac{R_{23}}{\delta_s - i} + \epsilon \left( \frac{R_{13}}{\delta_0 - \delta_s} - \frac{R_{23}}{\delta_0 - \delta_s - \delta_0 - i} \right) \left( \delta - \delta_s + i + \frac{\epsilon}{\delta_0 - \delta_s - \delta_0 - i} - \frac{\kappa}{\delta_0 + i} \right)^{-1} \right]
$$

$$
R_{13} = \{ R_{13}^0 [1 + 2(A_1 + V + S)] - R_{23}^0 (A_1 + V) - SR_{12}^0 \} Z^{-1}
$$

$$
R_{23} = \{ R_{23}^0 [1 + 2(A_0 + W + S)] - R_{13}^0 (A_0 + W) + SR_{12}^0 \} Z^{-1}
$$

where

$$
Z = 1 + 2(A_0 + A_1 + V + W + S) \tau + 3[A_0(A_1 + V) + S(A_0 + A_1 + V) + W(A_1 + V + S)] \tau^2.
$$

We have introduced here the dimensionless quantities

$$
\epsilon = \tilde{\epsilon} \tau^2 \quad \kappa = \tilde{\kappa} \tau^2 \quad \delta_0 = \Delta_0 \tau \quad \delta_s = \Delta_1 \tau
$$

for the sake of convenience in computer calculation of the quantities in (11).

### 3. Steady-state population distribution

In the limit of small Stokes intensity ($\kappa \ll \epsilon$, $\kappa \ll 1$) one obtains from equations (11c–d) the following expressions for the population differences:

$$
R_{13} = R_{13}^0 \frac{1 + \delta_0^2}{1 + \delta_0^2 + 4\epsilon} \quad R_{23} = R_{23}^0 - R_{13}^0 \frac{2\epsilon}{1 + \delta_0^2 + 4\epsilon}
$$


These are the well-known expressions for the population differences in a three-level system subjected to a strong pump field. The transition $1 \rightarrow 3$ is saturated ($R_{13} \rightarrow 0$). This saturation leads to inversion of the population difference $R_{23}$ ($R_{23} < 0$ for $\epsilon > [Q(1 + \delta_0^2)/2(1 - 2Q)]$ if $Q < \frac{1}{2}$, where $Q = R_{23}^0/R_{13}^0$.

We are interested now in the variation of the population distribution with increasing Stokes field.

The problem can be easily solved if both fields are in exact resonance. Then $\delta_0 = \delta_3 = 0$ and the steady-state populations are:

$$
\rho_{11} = \rho_{11}^0 \left( 1 - \frac{2\epsilon}{1 + 4\epsilon + 4\kappa (1 + 4\epsilon + 4\kappa)(1 + \epsilon + \kappa)} \right) + \rho_{22}^0 \frac{6\epsilon \kappa}{(1 + 4\epsilon + 4\kappa)(1 + \epsilon + \kappa)} + \rho_{33}^0 \frac{2\epsilon}{1 + 4\epsilon + 4\kappa}
$$

$$
\rho_{22} = \rho_{11}^0 \frac{6\epsilon \kappa}{(1 + 4\epsilon + 4\kappa)(1 + \epsilon + \kappa)} + \rho_{22}^0 \left( 1 - \frac{2\kappa}{1 + 4\epsilon + 4\kappa} \right) \frac{6\epsilon \kappa}{(1 + 4\epsilon + 4\kappa)(1 + \epsilon + \kappa)} + \rho_{33}^0 \frac{2\kappa}{1 + 4\epsilon + 4\kappa}
$$

$$
\rho_{33} = \rho_{11}^0 \frac{2\epsilon}{1 + 4\epsilon + 4\kappa} + \rho_{22}^0 \frac{2\kappa}{1 + 4\epsilon + 4\kappa} + \rho_{33}^0 \left( 1 - \frac{2\epsilon}{1 + 4\epsilon + 4\kappa} - \frac{2\kappa}{1 + 4\epsilon + 4\kappa} \right).
$$

Expressions (13) are attractive because of their simplicity and clarity and suggest that levels 1 and 2 are connected by two-photon transitions and the population transfers from levels 1 and 2 into 3 and vice versa are due to single-photon processes induced by strong resonance fields.

In figure 2 the steady-state populations of the levels are plotted as functions of the intensity of the Stokes wave ($\kappa$) for three fixed values of the exciting field (a) $\epsilon = 4$, (b) $\epsilon = 25$, (c) $\epsilon = 50$ for the system with initial populations $\rho_{11}^0 = 1$, $\rho_{22}^0 = \rho_{33}^0 = 0$ (i.e. $Q = 0$).

As it is seen from (13) for such a system at small $\kappa$ ($\kappa \ll \epsilon$), the levels 1 and 3 are mainly populated and the maximum population of level 3 can never exceed $1/2\rho_{11}^0$. With increasing Stokes field, the population of level 2 grows appreciably and for $\kappa = \frac{1}{2}\sqrt{(1 + 4\epsilon)(1 + \epsilon)}$, $\rho_{22}$ reaches a maximum value and then begins to decrease slightly. The ground state population considered as a function of $\kappa$ at fixed $\epsilon$ has a minimum for $\kappa \sim \epsilon/2$ and $\rho_{33}(\kappa)$ is a monotone decreasing function.

When the Stokes field becomes much stronger than the exciting field ($\kappa \gg \epsilon$) $\rho_{11} \rightarrow \rho_{11}^0$; $\rho_{22} \approx \rho_{33} \rightarrow \frac{1}{2}(\rho_{22}^0 + \rho_{33}^0)$, i.e. the transition 2–3 is saturated (analogously to the case $\epsilon \gg \kappa$ when the transition 1–3 is saturated).

In the case of a strong exciting field for values of $\kappa$, satisfying the inequality $0 < \kappa < \epsilon/2$, the intermediate level 3 is more populated than the ground and final states ($\rho_{33} > \rho_{11}$, $\rho_{22}$). If $\kappa$ falls in the range ($\frac{1}{2}\epsilon$, $\epsilon$), the population of the final level 2 is greater than that of levels 1 and 3 ($\rho_{22} > \rho_{11}$, $\rho_{33}$). As is seen, both conditions cannot be simultaneously satisfied. Furthermore, the inversion $\rho_{33} > \rho_{11}$, $\rho_{22}$ occurs for smaller pump fields compared with those arising from the inversion $\rho_{22} > \rho_{11}$, $\rho_{33}$ (figures 2(b), (c)).

Hence, the application of strong resonant fields on a three-level system with equal relaxation times can create the population inversion between excited and ground states. This result is a consequence of the simultaneous action of the two strong fields, inducing
two-photon transitions with probabilities equal to or greater than those of the single-photon transitions.

An analogous result has been obtained by Krainov (1976) for a three-level system without relaxations and by Whitley and Stroud (1976) for the case when the only damping mechanism in the system is the natural radiative decay. According to Whitley and Stroud (1976) the population of the excited level, connected with the ground level by two-photon transition, can be greater than 1/2. In the case of equal relaxation times considered, the maximum value of the excited level population cannot exceed 1/2.

The population inversion can be created not only under exact resonance conditions. In figure 3 the steady-state populations are plotted versus $\delta_\epsilon$ for $\kappa = 30$, $\epsilon = 50$ at resonance and off-resonance pumping respectively. One can see, with increased detuning of the exciting field, the region where $\rho_{22} > \rho_{11}$ shifts in direction and at a distance determined by the detuning $\delta_0$. It is interesting to note that in the case of
off-resonance excitation the inverse population between levels 2 and 1 arises at lower Stokes fields compared with the resonance case (e.g. for $\epsilon = 50$, $\kappa = 25$ at resonance pumping $\rho_{11} - \rho_{22} > 0$, while at off-resonance pumping the region where $\rho_{22} > \rho_{11}$ exists). On the other hand, for the same value of $\epsilon$ and $\kappa$ the population inversion is greater in some cases under off-resonance than under exact resonance conditions. This means that the resonance conditions are not the most effective conditions for obtaining population inversion between the ground and the excited levels. We shall come across a similar result below, considering the Stokes lineshape, where we shall see that in some cases the off-resonance conditions lead to a higher gain.

We should like to point out that the population inversion between the ground and the final excited level, obtained in this manner, is negligible. For that reason we think that this result is of academic rather than of practical interest.

If both applied fields are in exact resonance and their intensities are equal ($\epsilon = \kappa$) in the limit of strong fields the steady-state populations asymptotically approach the following values:

$$
\rho_{11} \rightarrow \frac{3}{8} - \frac{1}{8} \rho_{33}, \\
\rho_{22} \rightarrow \frac{3}{8} - \frac{1}{8} \rho_{33}, \\
\rho_{33} \rightarrow \frac{2}{8} + \frac{3}{8} \rho_{33}.
$$

(14)

In this limiting case the two-photon transition 1–2 is saturated.
4. The shape of the Stokes line

The imaginary part of the susceptibility $\chi''(\omega_s)$, which is responsible for attenuation or amplification at the frequency $\omega_s$, is given by the expression (11b).

In the weak Stokes signal limit, $\chi''(\omega_s)$ has the form:

$$\chi''(\omega_s) = N|d_23|^2 \hbar \text{Im} \left[ \frac{R_{23}}{\delta_s - i} + \epsilon \left( \frac{R_{13}}{(\delta_0+i)(\delta_s-i)} - \frac{R_{23}}{(\delta_0-i)^2} \right) \left( \delta_0 - \delta_s + i + \frac{\epsilon}{\delta_s-i} \right)^{-1} \right]$$

(15)

where $R_{ik}$ is given by (12).

A direct comparison of (15) and (11b) shows that the Stokes field increase leads to appearance of the term $\epsilon/(\delta_0+i)$ in the denominator. If we separate the real and the imaginary part of the denominator, i.e.

$$\left( \delta_0 - \delta_s + \frac{\epsilon \delta_s}{\delta_s+1} - \frac{\kappa \delta_0}{\delta_0+1} \right) + i \left( 1 + \frac{\epsilon}{\delta_s+1} + \frac{\kappa}{\delta_0+1} \right)$$

one sees that the frequency shifts caused by the applied fields $E_0$ and $E_s$ have opposite signs and hence they can compensate each other. On the other hand, the enhancement of the Stokes field leads to the broadening of the line (this is the so-called power-broadening effect).

These qualitative conclusions are confirmed by the results shown in figures 4–8 and discussed below as well as by the treatment by Panock and Temkin (1977).

We are interested in the power emitted or absorbed per unit length at a frequency $\omega_s$:

$$\frac{dI}{dz} = 2\omega_s E_s |\chi''(\omega_s)|^2.$$  

(16)

So we investigated $dI/dz$ as a function of $\delta_s$ for intensities of the Stokes field less than equal to and greater than the exciting field intensity ($\kappa \geq \epsilon$) for resonant ($\delta_0 = 0$) and off-resonant ($\delta_0 \neq 0$) pumping in the systems with an arbitrary equilibrium population distribution, i.e. $Q \neq 0$, where $Q = R_{23}/R_{31}$.

The case for $Q = 0$ investigated by Panock and Temkin (1977) is illustrated here mainly for comparison. It was interesting for us to distinguish the case $0 < Q < \frac{1}{2}$ and $\frac{1}{2} < Q < \frac{3}{4}$ because it is well known (Javan 1957, Kancheva 1977), that in the former case the amplification properties of the system are determined by direct emission as well as by the two-photon transition 1–2, while in the latter case no population inversion is established in the channel 2–3 and the amplification in such systems has a purely Raman-type origin.

First we shall consider resonant pumping when the exciting frequency is tuned to the centre of the line $\omega_0 = \omega_{31}$.

4.1. Resonant pumping

In figures 4(a)–(c) and figures 5(a)–(c) $dI/dz$ is plotted versus $\delta_s$ for several values of $\kappa$ and two fixed values of $\epsilon$ for the systems with the following population distribution: (a) $Q = 0$, (b) $0 < Q < \frac{1}{2}$, (c) $\frac{1}{2} < Q < \frac{3}{4}$.

One can see that for $Q = 0$ as well as for $Q \neq 0$ with increasing Stokes field the splitting disappears both in emission and absorption. After that, the maximum of $dI/dz$ is localised at the point $\delta_s = 0$. 
Figure 4. $(\tau/2\omega N)dl/dz$ is plotted versus $\delta_z$ for $\delta_0 = 0$, $\epsilon = 4$ and for various values of $\kappa$ for systems with population distribution (a) $Q = 0$, (b) $0 < Q < \frac{1}{2}$, (c) $\frac{1}{2} < Q < \frac{3}{2}$. 
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Figure 5. Stokes lineshapes for the same conditions as in figure 4 but for $\epsilon = 25$. 
From (11b) and (16) at $\delta_0 = \delta_s = 0$ one obtains:

$$dI/dz(\delta_s = 0) = 2\omega_0 N R_{13}^0 \frac{Q(1+2/\epsilon + \kappa) - 3\epsilon}{(1+\epsilon + \kappa)(1+4\epsilon + 4\kappa)} \kappa. \tag{17}$$

For $Q = 0$, (17) is equivalent to equation (14) from Panock and Temkin (1977). It is easy to calculate the value of $\kappa$ which maximizes $dI/dz$ for fixed value of $\epsilon$ in the case $Q = 0$. One obtains $\kappa = \frac{1}{2}(1+4\epsilon)(1+\epsilon)$ – this is the same value of $\kappa$ for which the population of level 2 attains its maximum. The value of $\kappa$, that maximises the power emitted in systems with $Q \neq 0$ can also be found by differentiating (17) with respect to $\kappa$, but the expression obtained is too cumbersome and will not be reproduced here. Both the calculated expression and figures 4 and 5 show that in systems with $Q \neq 0$ as a rule $dI/dz$ reaches its maximum for weaker Stokes fields than in the case $Q = 0$.

In a system with $0 < Q < \frac{1}{2}$ for a pump field inducing a well expressed doublet in emission (figures 4(b) and 5(b)), the behaviour of the emission lines with increasing $\kappa$ is analogous to that of the curves for population distribution $Q = 0$ (i.e. the emission enhances primarily, two peaks become intimate, the splitting disappears gradually, the amplification grows up to a given value of $\kappa$ and then decreases). But in contrast to the case for $Q = 0$ in which $dI/dz \rightarrow 0$ when $\kappa \rightarrow \infty$, now the emission line transforms into an absorption line for $\kappa > [3(3-4Q)/Q] - 1$. The absorption grows slowly with increasing $\kappa$ and for $\kappa \gg \epsilon$, $dI/dz$ asymptotically approaches the value $\omega_0 h N R_{23}^0/2\tau$ (saturation of absorption). In the course of this transition from emission to absorption, the system passes through a position in which, in the limits of the line $dI/dz = 0$, i.e. the system becomes transparent. So the system with $0 < Q < \frac{1}{2}$ can manifest itself as amplifying, attenuating or transparent, depending on the intensities of the exciting and Stokes fields.

When $\frac{1}{2} < Q < \frac{3}{4}$, the picture is different compared with those considered above. In such systems, when the Stokes field is a weak probe field and the exciting intensities exceed the value $\epsilon > Q/(3-4Q)$, a region of emission is induced in the vicinity of $\delta_s = 0$. The emission is accompanied by absorption regions on the left and right sides, i.e. in such systems (as well as in the case $Q > \frac{3}{2}$) the splitting appears in absorption. With increasing $\kappa$ both emission and absorption grow. Here, as well as in the cases discussed above, there exists a value of $\kappa$ maximising the emitted power. Above this value the emission decreases, but the enhancement and broadening of the regions of absorption continue. The two absorption peaks draw apart slowly, but the ‘valley’ between them gradually disappears as a result of the faster growth of $dI/dz$ in the region $\delta_s = 0$. For $\kappa \gg \epsilon$ the splitting disappears completely. The absorption line is too broad ($\gamma \sim 50$ for $\epsilon = 25, \kappa = 100$) with a maximum located at $\delta_s = 0$ and asymptotically approaching the value $\omega_0 h N R_{23}^0/2\tau$.

4.2. Off-resonant pumping

When the exciting field is off-resonant ($\delta_0 \neq 0$), the lines of emission (or absorption) are asymmetric. The fact that $\chi''(\omega_s)$ is symmetric about the point $\delta_s = \delta_0/2$ for $\delta_0 \neq 0$ (Panock and Temkin 1977) is not a general result for small Stokes intensity. It is true only for the case $Q = 0$. In the general case ($Q \neq 0$) for $\delta_0 \neq 0$ the curves $dI/dt$ versus $\delta_s$ are asymmetric both for weak and for strong Stokes signals.

In figures 6(a)–(c) and figure 7 the Stokes lineshape is presented for several values of $\kappa$ for $\epsilon = 4$ and 25 and $\delta_0 = 5$ in systems with (a) $Q = 0$, (b) $0 < Q < \frac{1}{2}$, (c) $\frac{1}{2} < Q < \frac{3}{4}$. 
Figure 6. \((\pi/2\hbar\omega_0 N)\,dI/dz\) is plotted versus \(\lambda\) for \(\delta_0 = 5\), \(\epsilon = 4\) and for various values of \(\kappa\) for systems with population distribution (a) \(Q = 0\), (b) \(0 < Q < \frac{1}{2}\), (c) \(\frac{1}{2} < Q < \frac{3}{2}\).
Here, as well as in the case of exact resonance, there is a value of $\kappa$ for which the amplification is maximal. The numerical computation of $dI/dz$ indicates that for off-resonant pumping the optimum gain in the systems with $Q = 0$ is the same as in the case of exact resonance, but if in the resonance case the gain reaches its maximum for $\kappa = \epsilon$, for $\delta_0 \neq 0$ this maximum is obtained for stronger Stokes fields. The greater the detuning from resonance, the stronger are the Stokes intensities for which $dI/dz$ has a maximum (compare figures 4(a), 6(a) and 8). If now the value of $\kappa$ exceeds $\kappa_m$ ($\kappa_m$ denotes the value of $\kappa$, which maximises $dI/dz$) for the same value of $\epsilon$ and $\kappa$, the amplification for $\delta_0 \neq 0$ is greater than for $\delta_0 = 0$. Consequently, as was noted in the previous section, the resonance excitation does not always lead to the optimum gain. Moreover, as one can see from figure 7 for the case $Q = 0.6$, the amplification is significantly greater for off-resonant than for resonant pumping. This fact is not surprising as in such systems (Kancheva 1977) the optimum gain is obtained under off-resonant rather than resonant conditions, when the Stokes field plays the role of a weak probe field.

It is interesting to reveal the behaviour of the lines in the systems with $0 < Q < \frac{1}{2}$ (see figure 6(b)). It is known in such systems for weak Stokes field and strong near-resonant exciting field, that simultaneous gain and absorption at different frequencies of the same line exist. With increasing $\kappa$, both emission and absorption enhance. They pass through a maximum and after that both decrease and on the right hand side of the region of emission, a second region of absorption forms. The new absorption band grows with increasing of $\kappa$ and for $\kappa \gg \epsilon$ the system appears solely in absorption. As is seen from the curves in figure 6(b), in such systems the enhancement of $\kappa$ leads to a smooth reverse of the spectrum. The absorption region situated on the short-wavelength side of the line for weak Stokes fields changes over to the long-wavelength side for strong Stokes fields.

Furthermore, considering the position of the emission peak of the curves of $dI/dz$ for the three cases (a), (b) and (c), it is seen that with increasing of $\kappa$ the peak moves from $+\delta_z$ to $-\delta_z$ passing through $\delta_z = 0$. What does this result yield practically?
Let us recall that for a weak Stokes field and a strong exciting field with positive detuning \((\Delta_0 = \omega_{31} - \omega_0 > 0)\) the Stokes Raman line undergoes a Stokes shift in comparison with the non-resonant Raman line (Kancheva 1977), the more clearly expressed, the stronger the pump field and smaller the frequency detuning \((\delta_s = \delta_{0/2} + \frac{1}{2}(\Delta_0^2 + 4\epsilon)^{1/2} \text{ for } Q = 0)\). Now, increasing \(\kappa\) at fixed \(\epsilon\) and \(\delta_0\), the emission peak moves from \(+\delta_s\) to \(-\delta_s\). Going over from the parameters \(\delta_s\) to the variables \(\omega_s = \omega_0 - \omega_{21} + (\delta_0 - \delta_s)\tau^{-1}\) it is seen that shifting of the peak means that the Stokes shift of the resonance Raman line is transformed to an anti-Stokes shift. For example: for \(Q = 0, \epsilon = 25, \delta_0 = 5\) and \(\kappa = 4, 25, 100\) the maximum of the emission line is located at \(\delta_s = 7.5, 5\) and \(0.5\) respectively.

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